RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 13 1 OCTOBER 1993-I

Interplay of superfluidity and phase separation in adsorbed films

M. Schick and O. E. Vilches
Department of Physics, FM-15, University of Washington, Seattle, Washington 98195
(Received 20 May 1993)

We note that when the line of superfluid transitions intersects a line of phase separation near the critical point of the latter, one should expect the simultaneous occurrence of both high- and low-density superfluids. The signals from a third-sound experiment depend distinctively upon the density, there being seven different regions of characteristic response, which facilitates the identification of such a possibility. This scenario may provide an explanation for recent observations on helium adsorbed on hydrogen, in which case the critical point would be that of a layer transition. We also describe the interesting progression which would be observed in the temperature–chemical potential plane as the superfluid line approaches such a point. This progression might be observable in microbalance experiments on the approach of the superfluid transition to the prewetting critical point.

There has been renewed interest recently in superfluidity in thin helium films. This has been due principally to the verification[1–6] of the suggestion by Cheng *et al.*[7,8] that helium will not wet the weak-binding substrates of alkali metals at very low temperatures. The implication is that, at liquid-gas coexistence, the helium does not form the usual thick Rollins film on the substrate, but only one of microscopic thickness. Furthermore, because the substrate is so weak, the film can be entirely in the liquid state in contrast to stronger binding substrates in which the first layer is invariably solid.

At some temperature below the three-dimensional liquid-gas critical temperature, the helium undergoes a first-order wetting transition at which the thickness jumps discontinuously from a microscopic to a macroscopic value, the latter being the thickness of the Rollins film.[9] In all the experiments cited above, the wetting temperature is below the bulk superfluid transition temperature. The discontinuity in thickness extends off of coexistence where it manifests itself as a jump from thin to thick films. The locus of such discontinuities is called the prewetting line. It ends at the prewetting critical point, a two-dimensional Ising critical point at which the jump in thickness goes to zero. The thick film becomes superfluid at the Kosterlitz-Thouless transition temperature,[10] $T_{\mathrm{KT}}(\mu)$, which depends upon the pressure or chemical potential. At this temperature, the areal density of superfluid, n_s, is discontinuous, with its value at a temperature infinitesimally below the transition given by[11]

$$\frac{n_s}{k_B T_{\mathrm{KT}}} = \frac{2m_{\mathrm{He}}}{\pi\hbar^2} . \tag{1}$$

Taborek and Rutledge[12] have recently carried out a series of measurements on helium adsorbed on a strong-binding gold substrate which was plated with several layers of weak-binding cesium. For a given cesium thickness, they follow the line of superfluid transitions as a function of pressure until this critical line terminates on the prewetting line at a two-dimensional critical end point [see Fig. 1(a)]. Thus, in these experiments, the thick film can be superfluid while the thin film cannot. The strength of the substrate can be increased by decreasing the thickness of the cesium film, and with this change the prewetting line becomes more parallel to the bulk coexistence curve and the wetting temperature decreases. It appears to vanish (occur at $T=0$) when the number of cesium layers is three or less. A similar increase in substrate strength and decease in wetting temperature can be brought about by employing a series of uniform, as opposed to plated, substrates of increasing adsorption strength, such as the sequence Cs, Rb, K, Na, Li, H_2, Mg considered by Cheng *et al.*[7,8] Because the increase in substrate strength decreases the wetting temperature at one end of the prewetting line, it is reasonable to expect that it also decreases the prewetting critical point at the other. This expectation is supported by calculation.[13] As a result, one can envision a scenario in which, by varying the substrate strength, the prewetting critical point

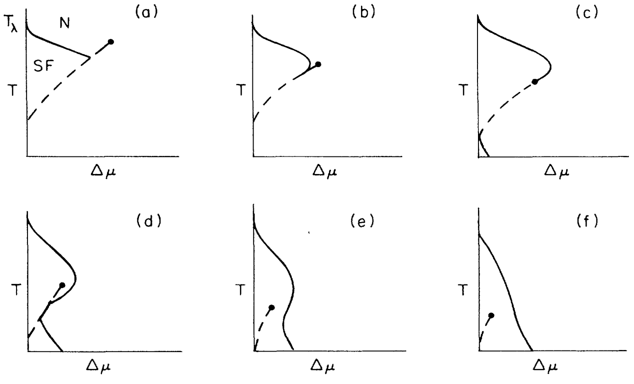

FIG. 1. Sequence of schematic phase diagrams in the chemical potential–temperature plane as the substrate potential is made stronger in going from (a) to (f). The line of superfluid transitions is shown solid, while the coexistence line is shown dashed. The critical point is shown as a dot.

0163-1829/93/48(13)/9910(3)/$06.00

passes from occurring above the superfluid transition line to occurring below it. It is this kind of scenario which we consider here. Our comments are not restricted to this particular scenario, but also apply to the interplay between the superfluid transition and other phase separations, such as layer transitions in thin films.

The points that we wish to make can be most easily illustrated in reference to Fig. 2. There we have shown schematically a phase separation in the temperature, areal-density plane, and the locus of superfluid transitions. We have chosen a case in which the critical point of the phase separation occurs just a little below the superfluid transition temperature at the critical density. If the superfluid density at the Kosterlitz-Thouless transition temperature $n_s\{T_{\mathrm{KT}}(n),n\}$ were a constant fraction of the total areal density n, then the locus of superfluid transitions would be a straight line in the T-n plane according to Eq. (1). However, as long as $n_s\{T_{\mathrm{KT}}(n),n\}$ is a smooth function of n, the form of the superfluid transition line will be similar to what we have shown, and our arguments are unaffected.[14]

Due to the fact that the coexistence curve has zero slope at the critical point, the superfluid transition line can intersect the low-density side of the coexistence curve, and will certainly do so if the temperature of the critical point, T_c, is sufficiently close to the Kosterlitz-Thouless transition temperature, $T_{\mathrm{KT}}(n_c)$, at the critical density n_c. Thus there are *two* coexisting superfluids in the temperature interval $T_c \geq T \geq T_1$, coexisting normal fluid and superfluid in the interval $T_1 \geq T \geq T_2$, and again two coexisting superfluids below T_2. (Note that the latter region of two-superfluid coexistence can exist even if $T_{\mathrm{KT}} < T_c$, while the former region cannot.) These various regions of coexistence have interesting, and distinct, signatures observable in experiments capable of detecting both superfluids simultaneously, such as those measuring third sound. The signatures from paths of constant density would depend upon the particular density interval sampled. Denoting the high- and low-density sides of the coexistence curve by $n_+(T)$ and $n_-(T)$ respectively, we can identify seven of them, as follows:

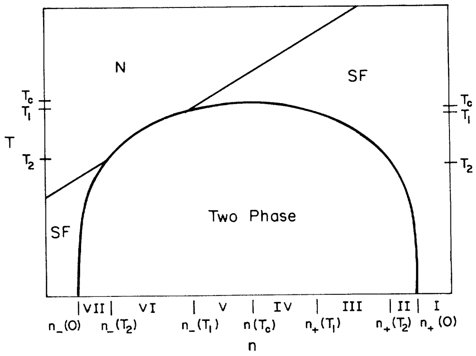

FIG. 2. Schematic phase diagram in the density-temperature plane showing a region of two-phase coexistence and a line of superfluid transitions which enters and exits the former at temperatures T_1 and T_2, respectively. The critical temperature is T_c. The normal and superfluid phases are labeled N and SF.

(I) $n > n_+(0)$. There is only one superfluid, and the onset of superfluidity is at $T_{\mathrm{KT}}(n)$.

(II) $n_+(0) > n > n_+(T_2)$. The onset is at $T_{\mathrm{KT}}(n)$. At lower temperatures, the phase boundary is encountered which gives rise to a change of slope in a plot of third-sound velocity vs temperature. At the same temperature at which this change of slope is encountered, a second third-sound signal appears corresponding to a less dense superfluid.

(III) $n_+(T_2) > n > n_+(T_1)$. The onset is at $T_{\mathrm{KT}}(n)$. At lower temperatures, the phase boundary is encountered which gives rise to a change of slope in a plot of third-sound velocity vs temperature. A second third-sound signal appears at the lower temperature T_2, one which corresponds to a lower density superfluid.

(IV) $n_+(T_1) > n > n(T_c)$. The onset is a $T_{\mathrm{KT}}(n)$. At a lower temperature, the phase boundary is encountered, evident in a change of slope of third sound velocity vs temperature. At the same temperature, a second third-sound signal appears, corresponding to a less dense superfluid. This signal disappears at T_1. It reappears at the lower temperature T_2.

(V) $n(T_c) > n > n_-(T_1)$. The onset is at $T_{\mathrm{KT}}(n)$. At a lower temperature, the phase boundary is encountered, evident in a change of slope of third-sound velocity vs temperature. At the same temperature, a second third-sound signal appears, one which now corresponds to a more dense superfluid. The original signal of the less dense superfluid vanishes at T_1, leaving only the signal from the more dense superfluid. At T_2, the signal from the less dense superfluid reappears.

(VI) $n_-(T_1) > n > n_-(T_2)$. The onset is no longer at $T_{\mathrm{KT}}(n)$, but occurs at the higher temperature at which the phase boundary is encountered. One signal appears there. Experimental plots of T_{KT} vs n_s should show a break from the straight line behavior of Eq. (1) in this region. At T_2, a second signal, corresponding to a low-density superfluid, occurs.

(VII) $n_-(T_2) > n > n_-(0)$. The onset is again at $T_{\mathrm{KT}}(n)$. At a lower temperature, the phase boundary is encountered at which a second signal appears corresponding to a higher density superfluid.

It should be noted that we have illustrated a case in which $n_-(0)$, the smaller of the two coexisting densities at zero temperature, is greater than zero. In a case in which it were zero, T_2 would vanish, and regions II and VII would not exist.

Two distinct third-sound signals have been reported recently[15] in systems of helium adsorbed on hydrogen and deuterium at coverages slightly in excess of one monolayer. The onset of the second signal appears to be correlated with a break in slope of the first signal, as in several of the above scenarios. It is possible that the signals arise from the interplay of the superfluid transition and a phase separation (layer transition) between one and two adsorbed layers. The interpretation of the results are made difficult due to the nonmonotonic relation between third-sound velocities and density.[16,17]

To conclude, we return to the question of the sequence of signals which should be observable as the critical temperature of phase separation, such as the prewetting critical point, is caused to pass through the superfluid transition line by varying an external parameter such as the substrate composition. We show in Fig. 1 the behaviors of the two transitions plotted in the temperature–chemical potential plane. When the line of superfluid transitions is caused to pass through the critical point of phase separation, it will approach that point along the direction of the temperaturelike variable and therefore will appear to be an extension of the coexistence line. This is shown in Fig. 1(c). When the superfluid transition line passes above the prewetting critical point, the former curves around the latter and intersects the coexistence line as shown in Fig. 1(d). Superfluidity will also appear at lower temperature at some point. It is a phase diagram like this one which is shown in the temperature-density plane in Fig. 2. Finally, as the prewetting critical point becomes further removed from the superfluid transition line, the gap between T_1 and T_2 of Fig. 2 disappears, and the superfluid transition line becomes unbroken [Figs. 1(e) and 1(f)]. The nature of the special multicritical point which occurs when the superfluid transition, in the universality class of the two-dimensional x-y model, intersects the phase separation critical point, in the universality class of the two-dimensional Ising model, is not clear. Because the Ising critical point and x-y transition lines exist for values of substrate potentials both stronger and weaker than the critical value at which the multicritical point occurs, it is tempting to believe that the two critical points are simply uncoupled from one another. More complicated scenarios are certainly possible, however.[18,19]

It would be very interesting to carry out experiments using *both* third-sound and microbalance techniques on either the cesium-plated gold or hydrogen substrates in order to monitor the interplay between superfluid and phase separation transitions. In the scenario that we have described, either set of signals is quite distinctive; taken together they should be able to determine clearly whether the coexistence of superfluid films we have described does indeed occur.

We wish to thank our colleagues J. G. Dash, M. P. M. den Nijs, and D. J. Thouless for instructive conversations. This work was supported in part by the National Science Foundation under Grant Nos. DMR 9220733 and DMR 9220729.

---

[1] P. J. Nacher and J. Dupont-Roc, Phys. Rev. Lett. **67**, 2966 (1991).

[2] K. S. Ketola, S. Wang, and R. B. Hallock, Phys. Rev. Lett. **68**, 201 (1992).

[3] P. Taborek and J. E. Rutledge, Phys. Rev. Lett. **68**, 2184 (1992).

[4] S. K. Mukherjee, D. P. Druist, and M. H. W. Chan, J. Low Temp. Phys. **87**, 113 (1992).

[5] E. Lerner, R. Morley, and O. E. Vilches, J. Low Temp. Phys. **89**, 593 (1992).

[6] J. E. Rutledge and P. Taborek, Phys. Rev. Lett. **69**, 937 (1992).

[7] E. Cheng, M. W. Cole, W. F. Saam, and J. Treiner, Phys. Rev. Lett. **67**, 1007 (1991).

[8] E. Cheng, M. W. Cole, W. F. Saam, and J. Treiner, Phys. Rev. B **46**, 13 967 (1992).

[9] For a recent review of wetting phenomena, see M. Schick, in *Liquids at Interfaces,* Proceedings of the Les Houches Summer School, Session XLVII, 1988, edited by J. Charvolin, J. F. Joanny, and J. Zinn-Justin (Elsevier, New York, 1991), p. 415.

[10] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973).

[11] D. R. Nelson and J. M. Kosterlitz, Phys. Rev. Lett. **39**, 1201 (1977).

[12] P. Taborek and J. E. Rutledge, Phys. Rev. Lett. **71**, 263 (1993).

[13] C. Ebner, W. F. Saam, and A. K. Sen, Phys. Rev. B **32**, 1558 (1985).

[14] As shown in Ref. 11, $n_s(T,n)$ for fixed n is a singular function of T as it approaches T_{KT} from below. However, we are concerned with the function of one variable, $n_s(T_{\mathrm{KT}}(n),n)$. There is no reason to believe that this is not a smooth function of n.

[15] M.-T. Chen, J. M. Roesler, and J. M. Mochel, J. Low Temp. Phys. **89**, 125 (1992).

[16] R. D. Puff and J. G. Dash, Phys. Rev. B **21**, 2815 (1980).

[17] E. Cheng, M. W. Cole, W. F. Saam, and J. Treiner, J. Low Temp. Phys. (to be published).

[18] E. Granato, J. M. Kosterlitz, J. Lee, and M. P. Nightingale, Phys. Rev. Lett. **66**, 1090 (1991).

[19] M. P. W. den Nijs, Phys. Rev. B **46**, 10 386 (1992).